

Restriction Requirement Issue

Applicants maintain the restriction requirement for the same reasons indicated in the Response filed September 16, 1996. The Examiner's argument that the catalytic system of the present invention can be used in other reactions besides a carbonylation process is without merit, because there would be no additional significant burden on the Examiner to search and examine the non-elected claims directed to a method for use of the same elected catalytic system claims 1-23 that the Examiner is presently examining. Reconsideration and withdrawal of this requirement are respectfully requested.

Issues Under 35 USC 103

Claims 1-23 have been rejected under 35 USC 103 as being unpatentable over EP 441446 (EP '446). This rejection is respectfully traversed for the following reasons.

Present Invention and Its Advantages

The present invention is directed to two catalyst systems (A) and (B) for carbonylation of an olefinically or acetylenically unsaturated compound. The catalytic system (A) comprises (A1) a Group VIII metal source of the Periodic Table of the Elements supported on a carrier, (A2) a ligand and (A3) an acid. The Group VIII metal (A1) may be cobalt, nickel, rhodium, palladium or platinum, or a compound of such metal. The carrier may be

activated carbon, an oxide of a metal or nonmetal oxide or a clay mineral. The ligand (A2) may be a phosphorus compound such as a tertiary organic phosphine, an arsenic compound such as a tertiary organic arsine or an antimony compound. The acid (A3) may be a protein (a Bronsted acid) such as an arylsulfonic acid, an alkylsulfonic acid, a carboxylic acid, a hydrohalogenic acid, sulfuric acid, a carboxylic acid, a hydrohalogen acid, nitric acid, a phosphoric acid or a perhalogenic acid.

The catalytic system (B) of the present invention for carbonylation comprises (B1) a Group VIII metal excluding palladium, (B2) a ligand shown by the following formula (Ib) and (B3) an electron donative compound having an election donability  $\Delta vD$  relative to a deuterate methanol material D of not less than 2:



wherein A represents a phosphorus atom, an arsenic atom or an antimony atom; and R<sup>1</sup>, R<sup>2</sup> and R<sup>3</sup> independently represent a hydrogen atom, an optionally substituted alkyl group, an optionally substituted alkenyl group, an optionally substituted alkynyl group, an optionally substituted cycloalkyl group or an optionally substituted aryl group, or R<sup>2</sup> and R<sup>3</sup> may together form an optionally substituted alkylene group, with a proviso that R<sup>1</sup> to R<sup>3</sup> are not

concurrently hydrogen atoms. As shown by the numerous Examples beginning at page 54 of the present specification, Applicants catalyst systems (A) and (B) insures a high transformation rate and excellent selectivity even after repeated use.

Distinctions Between Present Invention and EP '446

EP '446 (which corresponds to JP-4-215852 cited on page 2, lines 25-27 and on page 3, lines 1-9 of the specification) discloses a catalytic system comprising a Group VIII metal source, a phosphine having an aromatic substituent which contains an imino nitrogen compound, a proton source and a tertiary amine. EP '446 also discloses that the preferred Group VIII metal source is palladium. The EP '446 catalyst system is useful in the carbonylation of acetylenically and olefinically unsaturated compounds.

With respect to Applicants claimed catalytic system (A) comprising (1) a Group VIII metal source supported on a carrier (2) a ligand and (3) an acid, the EP '446 reference fails to disclose the use of a Group VIII metal catalyst supported on a carrier. EP '446 further directs that the use of the catalyst system may be homogenous or heterogenous, preferably homogenous (see page 6, lines 8-9). In all the Examples of EP '446, the catalyst system is used in a liquid phase. In contrast, the catalyst system (A) of the present invention uses a supported Group VIII metal catalyst in combination with a ligand and an acid. Therefore, the catalyst

system disclosed in EP '466 would not lead a person skilled in the art to use a supported Group VIII metal in combination with a ligand and an acid.

With respect to the catalyst system (B) of the present invention, Applicants catalyst (B) differs from EP '466 in that it is structurally different and specifically excludes palladium which is the preferred Group VIII metal in the EP '466 catalyst. Since the EP '466 catalyst prefers palladium, this reference clearly teaches away from Applicants invention. One important *indicia* of non-obviousness is "teaching away" from the claimed invention by the prior art. In re Bratt, 16 USPQ2d 1813 (Fed. Cir. 1990). In addition, the ligand compounds disclosed by EP '466 significantly differ in structure from Applicants' claimed ligand compounds. By definition the catalyst in EP '466 contains a heterocyclic ligand containing an aromatic substituent which contains an imino nitrogen atom. In contrast, Applicants' claimed catalyst system (B) contains a non-heterocyclic ligand that does not contain a nitrogen atom as a heteroatom. Therefore, to arrive at Applicants' catalyst system (B) based upon the EP '466 patent, one must not only modify the ligand compound but one must also exclude palladium as the Group VIII metal. The EP '466 patent fails to suggest to one of ordinary skill in the art the proposed required modifications to arrive at Applicants invention because the structure of Applicants' catalyst system B clearly differs from the catalyst system disclosed in EP '466.

In addition, the catalytic system disclosed in the EP '446 patent has significant disadvantages. Although it has an initial activity to some extent, it has a short catalyst life and thus is not suitable as a catalyst for commercial production. By way of illustration, when a homogenous catalyst system containing a palladium compound is used as a catalyst system, the palladium compound is reduced during the reaction and precipitates as a metallic simple substance, and the organic phosphine ligand is oxidized, which deteriorates the catalytic activity. Further, the EP '446 reference describes that the performance of triarylphosphine-based catalyst system is markedly impaired in the carbonylation of olefins when a tertiary amine is included. Therefore, in the EP '446 process using a tertiary amine such as pyridine, a phosphine having an aromatic substituent containing an amino-nitrogen atom is required to be used as the phosphine, and components for the catalyst system are remarkably restricted. Unexpectedly, however, Applicants have found that even when a Group VIII metal such as palladium is used in a catalytic system in combination with a ligand such as an organic phosphine (e.g. triarylphosphine) and an acid such as an alkyl sulfonic acid, supporting the catalyst on a carrier insures a high catalytic activity in spite of being a heterogenous catalytic system and moreover maintains such high catalytic activity even after repeated use.

Further, Applicants have found that a Group VIII metal, except

for palladium, such as platinum when used in combination with a specific ligand which does not contain a nitrogen atom as a heteroatom, and a specific electron donative compound does not decrease catalytic activity. Moreover, the use of such a catalytic system (B) rather ensures enhanced catalytic activity when employed in combination with an amine. Furthermore, the catalytic activity can be enhanced and a carbonylation product can be produced with higher selectivity.

Even if it is assumed that the disclosure of the EP '466 patent would render Applicants' catalytic systems (A) and (B) obvious, such *prima facie* obviousness is rebutted by the unobvious results obtained by Applicants. Applicants have demonstrated in the specification that favorable results are achieved by the claimed catalyst system (A) of the present invention comprising a Group VIII metal on a carrier, a ligand and an acid. The Examiner is referred to Applicants Examples 1-3 and Comparative Examples 1-3 respectively. As is apparent from Table 1 (Example 2, page 57), the use of the catalytic system (A) of the present invention in which a Group VIII metal source is supported on a carrier ensures a high transformation rate of 139 mol/hr, and excellent selectivity of 80%, even after the reaction procedure was repeated five times. To the contrary, the use of a palladium catalyst not supported on a carrier results in a significant decrease of the transformation rate and selectivity. After the reaction procedure was repeated five times using the palladium catalyst, the transformation rate

was only 53 mol/hr, which was almost one third of the initial rate and selectivity was at most 76%. The use of the catalytic system (A) of the present invention thereby ensures maintenance of a high transformation rate and excellent selectivity even after repeated use. These unexpected results are not suggested by the cited prior art.

With respect to the claimed catalytic system (B) comprising a Group VIII metal source excluding palladium, a ligand as defined in claim 1 and an electron donative compound, the Examiner is referred to Applicants' Example 5 and Comparative Example 5 in the specification (pages 60-62). In Comparative Example 5, a catalytic system comprising palladium (II) chloride, triphenylphosphine, methanesulfonic acid and pyridine was employed, and as a result of reaction, the transformation rate of propyne was only 3.7% and the selectivity for methylmethacrylate was only 52.7%. In contrast, the catalytic system (B) of Applicants' Example 5 in which a platinum compound was employed instead of a palladium compound showed a much higher transformation rate of 35.6% and a much higher selectivity of 96.9%.

Further, the Examiner is referred to Applicants' Example 30 and Comparative Example 8. As apparent from Applicants' Example 30, the use of Applicants' catalytic system (B) using a platinum compound according to the present invention results in excellent stability of the reaction system without precipitation of the catalyst as a metal. To the contrary, the use of the catalytic

system of Comparative Example 8 in which a palladium compound was employed in lieu of the platinum compound of Applicants' Example 30 resulted in deposition of most of the palladium thereby making the catalytic system extremely unstable.

The above unexpected results clearly rebut the alleged *prima facie* case of obviousness.

It is submitted for the reasons stated above that all of the presently pending claims define patentable subject matter such that the present application should be placed in condition for allowance.

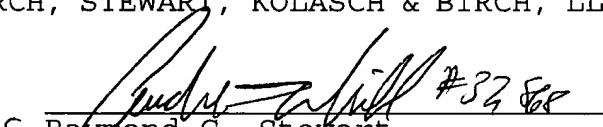
If the Examiner has any questions regarding the above matters, please contact Applicants representative Raymond C. Stewart in the Washington metropolitan area at the phone number listed below.

Please charge any fees or credit any overpayment pursuant to 37 CFR 1.16 or 1.17 to Deposit Account No. 02-2448.

Respectfully submitted,

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